Atmospheric phosphorus in the northern part of Lake Taihu, China

Jun Luo [a], Xiaorong Wang [a,*], Hong Yang [b], Jian Zhen Yu [b], Longyuan Yang [c], Boqiang Qin [c]

[a] State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing University, Nanjing 210093, China
[b] Department of Chemistry, Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China
[c] Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China

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ABSTRACT

Eutrophication, which has been linked to enhanced nutrient loading in lakes, estuarine and coastal waters, is common to many water bodies in China, including Lake Taihu. The atmosphere is an important and growing source of new nutrients entering water bodies. To assess the impact of atmospheric phosphorus (P) on water quality of Lake Taihu, we have estimated the atmospheric input of P by measuring its concentrations in aerosols and in rainwater. Samples were collected at the northeastern part of the lake. The P concentration in total suspended particle ranged from 0.054 to 0.300 $\mu g\ m^{-3}$. Concentrations of P in aerosols in the winter and autumn samples were higher than those in the spring and summer samples. Soluble inorganic P (SIP), which provides direct nutrient sources to algae, accounts for 4.9%–83.4% of the total P. Principal Component Analysis of the measured aerosol constituents and examination of inter-species correlation indicate that SIP was correlated with elemental carbon, water soluble organic carbon, NO_3^-, and Ca^{2+}. Such correlations could be explained if two nearby coal-fired power plants were a major aerosol P sources. The average concentration of the total P in rain water samples was 0.082 ± 0.058 mg P/L. The annual deposition flux of P into the northern part of Lake Taihu was estimated to be in the range of 0.04–0.44 kg ha^{-1} $year^{-1}$ through dry deposition and 0.33 kg ha^{-1} $year^{-1}$ through wet deposition. The combined wet and dry deposition was less than 8% of riverine P input, therefore the atmospheric deposition was only a minor contributor to P in Lake Taihu at the time of this study.

1. Introduction

Lake Taihu (30°5′–32°8′N, 119°8′–121°55′E) (Fig. 1), with an area of 2338 km², is the third largest freshwater lake in China (Qiu et al., 2008). It supplies drinking water and other water uses for more than 34 million people around the lake. The last two decades have seen fast economic development in the Lake Taihu drainage basin. This has exacted a toll on the water quality of Lake Taihu. The lake has become highly eutrophicated. Harmful algal blooms (HABs) have become a common phenomenon in Lake Taihu and have occurred every year in the past decades (Shen et al., 2001; Zhou and Zhu, 2003; Xu et al., 2010). Eutrophication and HABs have been linked to enhanced nutrient loading in lakes, estuarine and coastal waters (Pollman et al., 2002).

The atmosphere is an important and growing source of new nutrients entering many water bodies. Studies on coastal waters in North America and Western Europe indicate that from 20 to >40% of biologically available new nitrogen (N) inputs into coast-

al waters are of atmospheric origin (Paerl and Whitall, 1999). In the past, atmospheric fluxes of phosphorus (P) have generally been ignored because of a lack of data (Stumm, 1973; Lerman et al., 1975; Chen et al., 1985). There are reports showing that the atmospheric input of nutrient P amounts to about 10% of riverine input for some lakes (Duce, 1986; Moutin et al., 1998). To understand the nutrient cycling and to assess its impact on water quality, we need to quantify the atmospheric input of P and identify its main sources.

Lake Taihu is the most-studied lake in China. There are more than 10 years of observational data on water quality (e.g., nutrient concentrations, chlorophyll concentrations, dissolved oxygen, and etc.), meteorological data and hydrodynamic measurements. Numerous scientific studies of Lake Taihu have also been published, providing detailed insights into various environmental processes that contribute to the water quality problem in Lake Taihu (Chen et al., 2003; Zhang et al., 2006). In contrast to a wealth of information on the aqueous system, the amounts and chemical forms of N and P input from the atmosphere to Lake Taihu are largely unknown. We have carried out a yearlong study to collect atmospheric samples in a location in the northeast part of Lake Taihu. In this paper, we report concentrations

* Corresponding author. Tel.: +86 25 83595682; fax: +86 25 83595222.
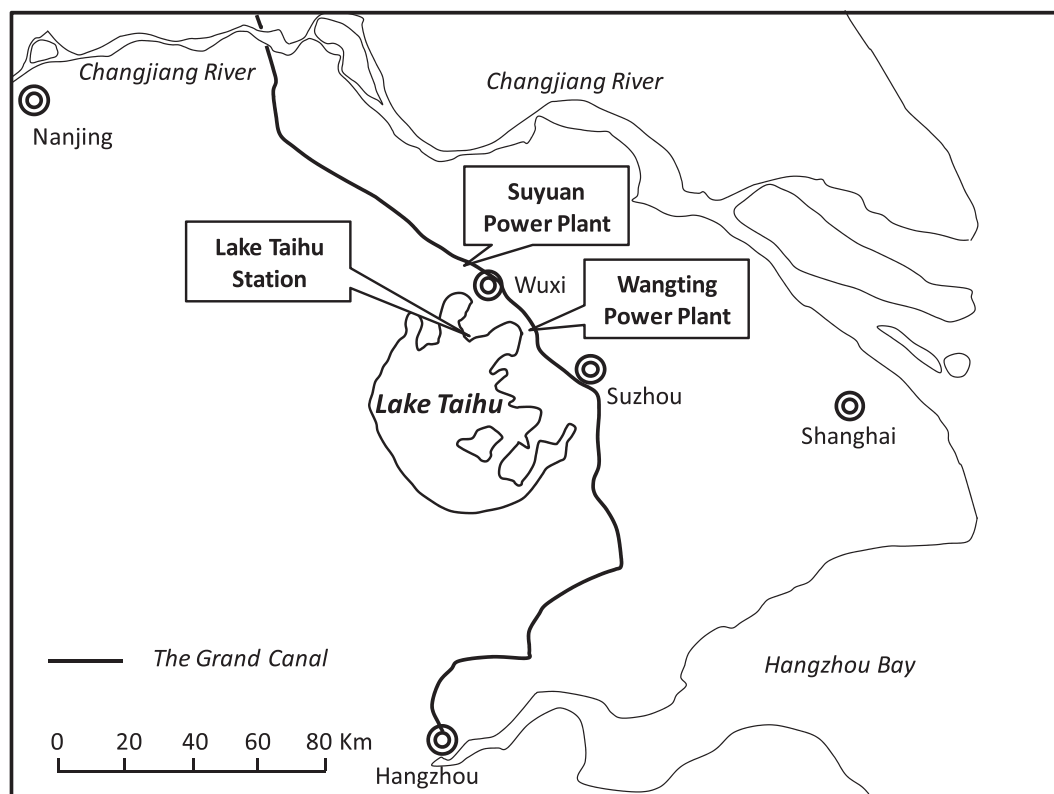E-mail address: ekxr@nju.edu.cn (X. Wang).

Fig. 1. The topographical map of Lake Taihu and the sampling location.

of several P forms in aerosols and total P concentrations in rain water samples. The aim of this study is to estimate input of atmospheric P by wet and dry deposition into Lake Taihu.

2. Experiment

2.1. Sample collection

Aerosol samples were collected at a field station (31.40°N, 120.22°E) (Xu et al., 2010), located in the northeast part of Lake Taihu (Fig. 1). The station is one of the 29 field stations in Chinese Ecosystem Research Network. The nearest suburban center is 3 km to the north. The aerosol sampler, a Hi-vol total suspended particle (TSP) sampler (Andersen, USA), was placed on a raised platform of a diameter of forty meters. Aerosol phosphorus was collected onto a quartz filter (8 × 10 inch) at a flow rate of ~700 L min^{-1} for 24 h. The filters were combusted at 550 °C overnight to remove any absorbed organic materials, then wrapped in annealed aluminum foil pouches before use. Samples were obtained in the months of Feb/Mar, Apr/May, Jul/Aug, and Oct/Nov in 2003 to represent winter, spring, summer, and autumn, respectively. In each sampling month, one sample was collected every 4 d. A field blank was collected in each sampling month. All the filter samples were stored at −4 °C in a freezer until analysis (Yang et al., 2005).

Rainwater was collected in a 3 L glass beaker at the same site during each rain event from August 2002 to July 2003. A total of 56 rainwater samples were collected. The rainwater amount was measured using a rain gauge. The beaker was placed on a platform 2 m above the ground. Before sampling, the beaker was soaked in a 10% HNO₃ solution for 24 h, followed by rinsing with ultra pure water. Immediately after collection, the rainwater sample was transferred to a polyethylene bottle without filtration and stored at −10 °C until analysis. Rainwater analysis was carried out at the end of each month.

2.2. Sample analysis

2.2.1. Determination of various forms of phosphorus

Separate circular segments of 2.24 cm in diameter were removed from the 8 × 10 in filter for analysis of soluble inorganic phosphorus (SIP) and total phosphorus (TP). The two types of P, after various pretreatments, were measured by a molybdenum blue technique with colorimetric detection (Murphy and Riley, 1962). Fig. 2 outlines the sample treatment procedures for various P species. The analysis of SIP followed the procedure described by Murphy and Riley (1962). One segment was soaked and extracted by sonication in 10 ml of ultra pure water in a clean, sealed polyethylene tube for 30 min before analysis (Chen et al., 1985).

Three different methods were applied to determine TP (Murphy and Riley, 1962; Graham et al., 1979; Migon et al., 2001). The first TP method involves the use of HNO₃ and HF to digest the filter samples. In this method, a filter segment of 2.24 cm in diameter was placed in 5 ml of 65% HNO₃ in a Teflon flask. The flask was closed and placed on an electrothermal board at 60 °C for 4 h and then at 150 °C for 4 h until only dry residue remained. The flask was cooled and left open in a fume hood until the acid fumes dissipated. The dry residue was then re-dissolved in 2 ml 65% HNO₃ and 3 ml 40% HF. The sample was heated on the electrothermal board at 60 °C for 3 h and subsequently at 150 °C for 4 h until a white dry residue was obtained. This residue was dissolved in 10 ml of 1 M HCl and ready for colorimetric analysis (Migon et al., 2001). A second method to determine TP was to ash a filter segment at 550 °C for 1 h followed by dissolution of the ash residue in 10 mL of 1 M HCl under sonication for 30 min (Graham et al., 1979). In the third TP method, a filter segment was treated with 8 mL 10% potassium persulphate solution, then was autoclaved for 30 min at 120 °C followed by colorimetric determination of phosphate (Menzel and Corwin, 1965). TP determined using these three methods are designated as TP$_{HNO_3}$, TP$_{ashed}$, and TP$_{persulfate}$, respectively, in ensuing discussions.
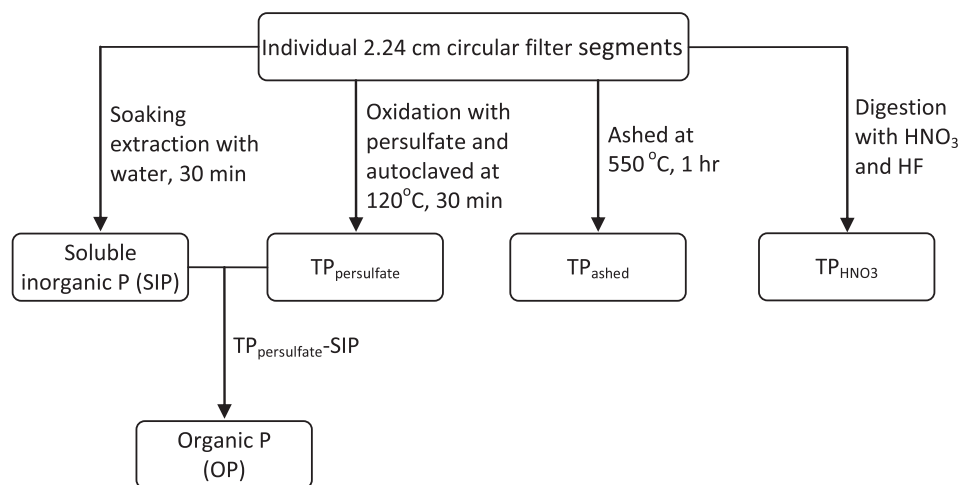
Fig. 2. Treatment flow chart for determination of various P Species.

Two forms of P, organic P (OP) and refractory P (R_fP), do not have direct methods for their measurements. They were calculated from other P measurements. OP was taken to be the difference between $TP_{persulfate}$ and SIP (Chen et al., 1985). R_fP was defined as the difference between TP_{HNO_3} and $TP_{persulfate}$.

TP in rainwater samples was determined using the persulfate-digestion method. An aliquot of 6 mL rain water was treated with 4 mL 10% potassium persulphate solution, autoclaved for 30 min at 120 °C followed by colorimetric determination of phosphate (Menzel and Corwin, 1965).

2.2.2. Other chemical analyses

Soluble inorganic ions (NO_3^-, SO_4^{2-}, Cl^-, Ca^{2+}, Na^+, Mg^{2+}, NH_4^+) and oxalate in the water extracts of the TSP samples and soluble F^- in rainwater samples were determined using an Ion Chromatographic system (DX-300, Dionex).

Elemental carbon (EC) and water soluble organic carbon (WSOC) were measured using a thermal/optical aerosol carbon analyzer (Sunset Laboratory, Forest Grove, OR, USA) (Yang et al., 2005). A filter segment of 1.45 cm^2 in size was removed from the 8 × 10 in filter and loaded into the aerosol carbon analyzer. The detailed information on the thermal analysis condition has been published elsewhere (Schauer et al., 2003). All the EC analyses were in duplicate.

As for the measurement of WSOC, 20 filter pieces (1.45 cm^2 in size) were extracted with ultra pure water in a sonication bath for 15 min (Yang et al., 2003). After filtration the water extracts were concentrated using a rotary evaporator to 0.5 ml followed by analysis using the carbon analyzer with the same thermal analysis conditions as those for EC measurement. The concentration of WSOC in water extract of the blank filters was 0.43 ± 0.08 μg ml^{-1}, at least 10 times lower than the samples.

2.3. Data analysis

Correlation plots were obtained using Microsoft Excel. The probability levels of the correlations were performed using SPSS 16.0 (SPSS, IL.). Principal components analysis was performed using XLSTAT (Addinsoft Inc., NY.).

3. Results and discussion

3.1. Method characterization

For the colorimetric analysis, the limit of detection (LOD) is 0.02 mg L^{-1}. Here the LOD was calculated as three times the standard deviation of the blank (n = 6). The analytical accuracy of the colorimetric analyses was assessed by conducting recovery checks. The recovery experiments of various P species were conducted by adding 0.5 mL of 2 μg L^{-1} KH_2PO_4 standard solution. The recoveries were 102.4 ± 4.4% for SIP, 95 ± 0.5% for TIP, 85.3 ± 3.2% for TP_{ashed}; 100.5 ± 3.7% for TP_{HNO_3}, and 91.5 ± 1.4% for $TP_{persulphate}$.

Due to limited aerosol samples collected in the summer, the measurement of TP using ashing method was not performed on the summer samples. The TP measurements of 19 samples in the other three seasons by the three TP methods are compared. It is found that the three methods are in agreement within 15%, with the HNO_3 digestion method generally yielding the highest TP and the ashing method giving the lowest TP (Fig. S1 in the Supplementary materials). In the ashing method, some of P in samples becomes vaporized as P_2O_5, which vaporizes at 300 °C (Newman, 1995). This explains why the TP concentrations measured by the ashing method gave the lowest values in general. In the acid digestion method, the combination of HNO_3 and HF could destroy the solid lattice of the aerosol particles and release some refractory P, which was not released in the potassium persulphate method.

### 3.2. Air mass back trajectory analysis

The sampling site is under the influence of southeast–northwest monsoon winds. The origin of air masses during each sampling month was analyzed through 10 d back trajectories using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model from National Oceanic and Atmospheric Administration (NOAA) (available at http://www.arl.noaa.gov). The back trajectories were computed at 500 m above sea level. The back trajectory analyses show that the air masses were transported to the sampling site over inland of the Asian Continent (China, Mongolia and Russia, etc.) in winter, spring and autumn and through the northwest Pacific Ocean in summer (Fig. S2).

3.3. Concentrations of phosphorus in aerosols above Lake Taihu and its possible sources

Generally the percentage of SIP varied from 4.9% to 83.4% of TP_{HNO_3} and TIP accounted for about 52% of TP_{HNO_3}. OP had a range of 11.4–87.8% of TP_{HNO3}. R_fP was a minor fraction in TP_{HNO_3}, accounting for 0.7–34.1% of TP_{HNO_3}. Concentrations of various P species in aerosols are presented in Table 1 and Fig. 3. The average concentrations of TP in winter and autumn were higher than that in spring and in summer. This is a combined result of influence of precipitation and air mass origins. Summer and spring had more precipitation and therefore more removal of aerosol by wet

Table 1
Concentrations of various species of aerosol phosphorus over the northern part of Lake Taihu (unit: $\mu g/m^3$).

ID		Sampling date M/D/Y	Total P			Various P		
			TP_{HNO_3}	TP_{ashed}	$TP_{persulfate}$	SIP	OP	R_fP
Winter	1	02/27/2003	0.161	0.172	0.159	0.045	0.114	0.002
	2	03/03/2003	0.227	0.122	0.192	0.035	0.157	0.035
	3	03/07/2003	0.245	0.176	0.209	0.103	0.106	0.036
	4	03/11/2003	0.129	0.036	0.119	0.006	0.113	0.010
	5	03/15/2003	0.091	0.075	0.072	0.022	0.050	0.019
	6	03/19/2003	0.300	0.244	0.198	0.065	0.133	0.102
	7	03/23/2003	0.157	0.145	0.144	0.052	0.093	0.013
	Mean		0.174	0.139	0.156	0.047	0.109	0.031
	Standard deviation		0.086	0.069	0.049	0.031	0.033	0.034
Spring	8	04/14/2003	0.158	0.137	0.145	0.047	0.098	0.013
	9	04/18/2003	0.105	0.087	0.104	0.044	0.060	0.001
	10	04/22/2003	0.142	0.149	0.137	0.059	0.078	0.006
	11	04/26/2003	0.124	0.093	0.108	0.043	0.064	0.016
	12	04/30/2003	0.150	0.133	0.139	0.077	0.063	0.011
	13	05/04/2003	0.176	0.132	0.155	0.043	0.111	0.021
	Mean		0.142	0.122	0.131	0.052	0.079	0.011
	Standard deviation		0.025	0.025	0.021	0.013	0.021	0.007
Summer	14	07/14/2003	0.058		0.055	0.047	0.008	0.002
	15	07/18/2003	0.056		0.052	0.036	0.016	0.003
	16	07/22/2003	0.059		0.057	0.048	0.008	0.002
	17	07/26/2003	0.054		0.046	0.040	0.006	0.008
	18	08/03/2003	0.072		0.068	0.055	0.012	0.004
	19	08/07/2003	0.082		0.081	0.068	0.013	0.001
	Mean		0.064		0.060	0.049	0.011	0.003
	Standard deviation		0.011		0.013	0.011	0.004	0.003
Autumn	20	10/17/2003	0.276	0.230	0.249	0.177	0.072	0.026
	21	10/25/2003	0.285	0.237	0.266	0.187	0.078	0.019
	22	10/29/2003	0.186	0.150	0.182	0.084	0.098	0.004
	23	11/02/2003	0.288	0.262	0.262	0.191	0.070	0.027
	24	11/06/2003	0.158	0.093	0.150	0.083	0.068	0.007
	25	10/09/2003	0.134	0.088	0.126	0.072	0.054	0.008
	Mean		0.221	0.177	0.206	0.132	0.073	0.015
	Standard deviation		0.070	0.076	0.061	0.058	0.014	0.010

SIP, soluble inorganic phosphorus; OP, organic phosphorus; R_fP: refractory phosphorus.
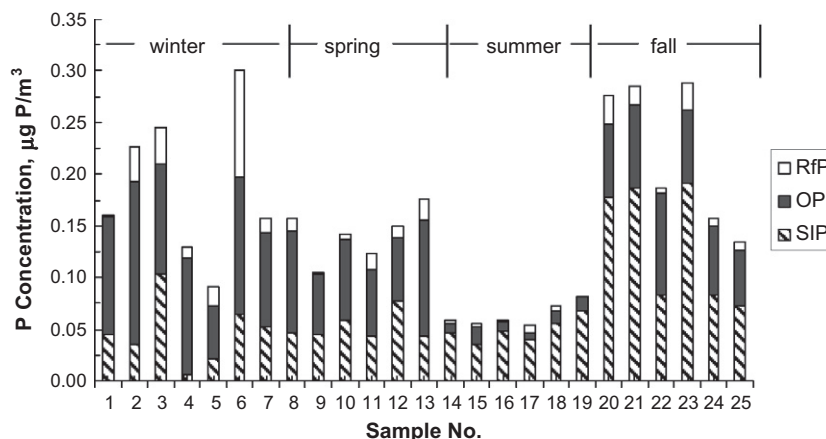
Fig. 3. The concentrations of various species of phosphorus in aerosols above Lake Taihu. (R_fP: residue phosphorus; OP: organic phosphorus; SIP: soluble inorganic phosphorus).

deposition. The generally more polluted air masses transported from northern China in winter and autumn contributed to the high loadings of aerosol P. The highest SIP was observed in the fall while the highest OP and R_fP were observed in the winter. The out of sync seasonal variation in SIP and OP indicates that the two forms of P derived from different sources. OP and R_fP were weakly correlated ($R^2 = 0.29$). The limited number of samples collected in each season made it difficult to extract information on the inter-relationships between different forms of P.

Table 2 summarizes previous measurements of aerosol phosphorus concentrations published in the literatures. Concentrations of aerosol TP above Lake Taihu were from 0.054 to 0.300 $\mu g\ m^{-3}$. It is obvious that aerosol P concentrations above Lake Taihu were the highest among the measurements, indicating stronger aerosol P sources in this region.

Principal Component Analysis (PCA) of the measured aerosol constituents yielded two principal components (PC), accounting for 83% of the total variance of the data set (63% and 20% by the first and the second PC, respectively). Table 3 lists the factor loadings of the input variables in PC1 and PC2. In PC1, SIP, EC, OC, WSOC, Ca^{2+}, NH_4^+, K^+, SO_4^{2-}, and NO_3^- have high loadings and Cl^- and Mg^{2+} have moderate loadings, signifying the association of this

Table 2
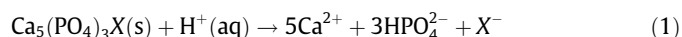Comparison of aerosol total phosphorus over Lake Taihu and at other sites previously studied.

Location	Site characteristic	Aerosol type	TP ($\mu g\ m^{-3}$)	No. of samples	Analysis method	Citation
Lake Taihu	Rural surrounding in a region of fast economic development and urbanization, rural	TSP	0.054–0.300	25	Digested with HNO_3 and HF	This work
Western Mediterranean Sea	Southeastern coast of France, rural	TSP	0.025–0.085	48	Digested with HNO_3 and HF	Migon et al. (2001)
New York, USA	New York city, urban		0.1–0.13			Graham et al. (1979)
Amazon	The Ducke forest reserve, rural	5-stage impactors	0.017	60	X-ray emission	Artaxo and Hansson (1995)
Corsica, France	Rural		0.010–0.020			Bergametti et al. (1992)
New Zealand	90 mile Beach on the northwest coast of North Island of New Zealand, rural	TSP	0.004	20	Sample was ashed at 500 °C	Chen et al. (1985)
South Africa	The aerosol monitoring network in the power generating region on the south African Highveld	TSP	0.02–0.08		Ion chromatography	Scheifinger and Held (1997)
Connecticut, USA	East of the Connecticut river valley, and along the Connecticut coast of the Long Island Sound, rural	Three-stage air filterpacks	0.01		Extraction	Yang et al. (1996)

factor with both a major EC source and the secondary components of the aerosols and SIP is mainly associated with this factor. In PC2, OP, Na^+, and Cl^- have high loadings, indicating that OP is associated with sea salt aerosol source. Chen et al. (1985) also found that the OP in aerosols over northern New Zealand is from the sea.

Correlations of concentrations of SIP with the measured major aerosol constituents were also examined. Positive correlations ($R^2 > 0.58$, $n = 25$, $p = 0.01$) were found between SIP and EC, WSOC, nitrate, sulfate, oxalate, and soluble Ca (Fig. 4), confirming the PCA results. EC is known to derive from combustion of carbonaceous fuels (e.g., coal, petroleum); nitrate, sulfate, oxalate, and WSOC are secondary aerosol components (Yang et al., 2005); and Ca^{2+} is associated with dust particles and acid dissolution processes.

It is possible that coal burning was the dominant source for EC, SIP and Ca^{2+}. Two coal-fired power plants, Wangting power plant and Suyuan power plant, are located approximately 25 km to the east and north of the sampling site, respectively (Fig. 1). Coal contains P mostly in the form of apatite and other complex phosphates. These P forms do not vaporize during coal combustion and as a result become part of fly ash (Newman, 1995). Ca is also a component of fly ash, mainly exist in its oxide form (i.e., calcite) (Ugurlu and Salman, 1998). Fly ash particles vary in size from some large enough to fall within a few kilometers of firepower plants to

Table 3
Factor loadings of two factors identified by Principal Component Analysis of the measured aerosol constituents.

	F1	F2
SIP	**0.908**	
OP	0.267	**0.811**
EC	**0.839**	0.247
OC	**0.959**	
WSOC	**0.924**	
Ca^{2+}	**0.851**	*−0.318*
Na^+	0.277	**0.851**
NH_4^+	**0.861**	
K^+	**0.919**	
Mg^{2+}	**0.707**	*0.589*
SO_4^{2-}	**0.857**	−0.271
NO_3^-	**0.926**	
Cl^-	**0.669**	**0.668**
Oxalate	**0.757**	−0.460

Note: Bold and italicized values represent factor loadings higher than 0.60 and 0.4–0.6, respectively. Factor loadings less than 0.23 are omitted.
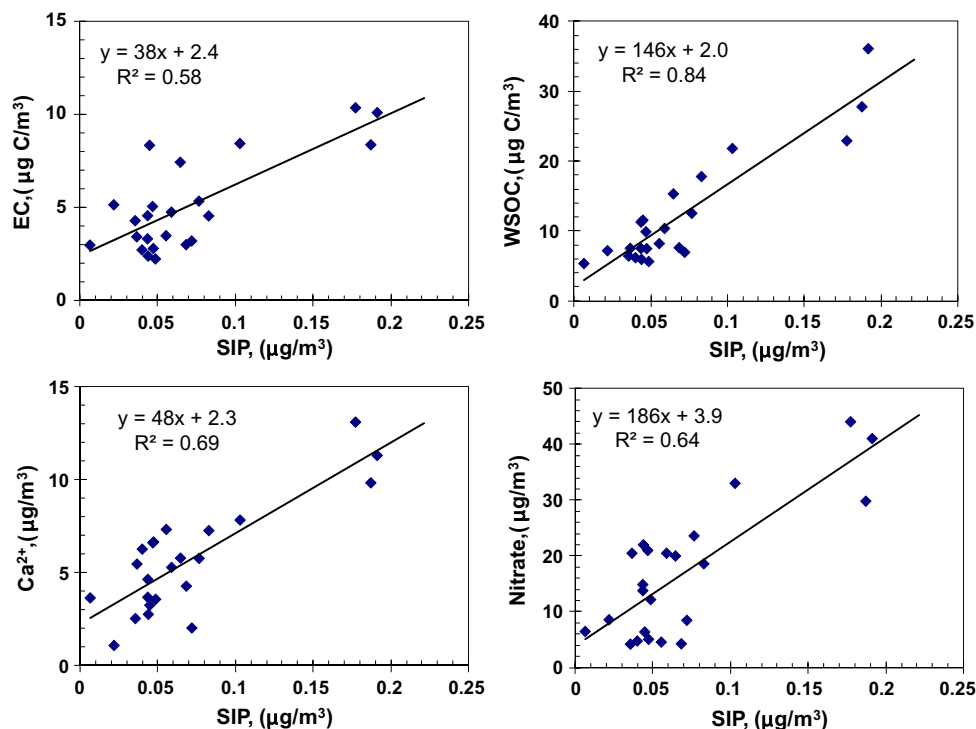
fine smoke particles traveling thousands of kilometers (Aitken et al., 1984; Alderliesten et al., 1987). It is expected that P and Ca, when freshly emitted, are largely insoluble. As fly ash particles are transported away from their sources, acidic species (e.g., H_2SO_4 and HNO_3) are adsorbed onto their surface, leading to dissolution of both Ca and insoluble P.

$$Ca_5(PO_4)_3X(s) + H^+(aq) \rightarrow 5Ca^{2+} + 3HPO_4^{2-} + X^- \qquad (1)$$

In the above chemical equation, $Ca_5(PO_4)_3X$ (s) represents apatite and X represents F^-, Cl^-, and OH^-. H^+ is from the strong acids in atmosphere, such as sulfuric acid and nitric acid.

Additional evidence for the influence of coal burning came from F^- measurements in rainwater samples. The annual mean concentration of F^- in rainwater samples in Lake Taihu was 0.327 mg L^{-1}. This level was much higher than those typically found in other places outside China, but lower than measurements made in Chengdu (Table S2). Chinese coal has been reported to have fluorine content of ∼200 mg kg^{-1}, twice as high as the mean level of 100 mg kg^{-1} found in coals elsewhere (Chen and Tang, 2002). The high F^- concentrations in the rainwater samples were consistent with the suggestion of significant influence of coal burning on the aerosols above Lake Taihu.

3.4. Estimation of atmospheric input of Phosphorus to Lake Taihu

Persulfate releasable phosphorus ($TP_{persulfate}$) was used as the total phosphorus to calculate atmospheric P input to Lake Taihu for the following reasons: (1) TP_{HNO_3} also includes recalcitrant inorganic P within silica and this part cannot be readily released to the aqueous environment (Newman, 1995); (2) TP_{ashed} may underestimate the actual P due to volatile loss during analysis.

The atmospheric deposition flux of P (F, $\mu g\ (m^2\ d)^{-1}$) can be estimated from the aerosol concentrations (C, $\mu g\ m^{-3}$) by multiplying the velocity of dry deposition (V, cm s^{-1}) (Migon et al., 2001) as follows:

$$F = C \times V \qquad (2)$$

This approach has been used in a number of investigations, including the Clean Air Status and Trends Network (CASTNet) in the US. The velocity of dry deposition is controlled by many factors (Junge, 1963; Duce et al., 1991) and can be given by $V_t = V_s + V_i$, where V is the rate of deposition and subscripts t, s and i refer to

Fig. 4. Plots of soulable inorganic P (SIP) data vs. Ca^{2+}, WSOC, NO_3^- and EC concentrations.

total, sedimentation, and impaction, respectively. Duce et al. (1991) suggested using a mean V value of 0.1 cm s^{-1} for aerosols of <1 μm and 1.0 cm s^{-1} for aerosols of >1 μm. The P concentrations in TSP determined in this study includes P species that reside on particles of various sizes. While SIP was likely associated with coarse particles, we had no knowledge of the size distributions of other P species. Consequently, we estimated the dry deposition of P using both V values to obtain an upper and lower limit estimates.

Table 4 shows the estimated seasonal and annual average dry flux estimates of aerosol P. On an annual average basis, the lower limit estimate was 12 μg (m^2 d)$^{-1}$ while the upper limit estimate was 120 μg (m^2 d)$^{-1}$. If these estimates are applied to the northern part of Lake Taihu, with an area of 120 km^2, the annual amount of P deposited in the lake due to dry deposition is 0.04 kg ha^{-1} year^{-1} if we assume a deposition velocity of 0.1 cm s^{-1} and 0.44 kg ha^{-1} year^{-1} if we assume a deposition velocity of 1.0 cm s^{-1}.

The wet deposition flux was also estimated from the measurements of TP in the rainwater samples. The annual average concentration of TP in rain water was 0.082 ± 0.058 mg P/L. The wet deposition input flux of P was then calculated to be 0.33 kg ha^{-1} year^{-1}. This puts the combined wet and dry deposition in the range of 0.34–0.77 kg ha^{-1} year^{-1}. The wet deposition flux of P falls between the lower and the upper estimate of P dry deposition. If we choose the median value of dry deposition, the

P fluxes of wet deposition and dry deposition are of a comparable magnitude.

The atmospheric deposition of P was small in comparison with the riverine input. The main pollution input into Lake Taihu is from river (Xu et al., 2010). There are four main riverine sources of P emptying into the northern part of Lake Taihu (Yu et al., 2010). The total riverine P input to the northern part of Lake Taihu was estimated to be 286 t y^{-1} (24 kg ha^{-1} year^{-1}) in 1995 (Liu et al., 1998) and 109 t y^{-1} (9.1 kg ha^{-1} year^{-1}) in 2003 (Huang et al., 2005). The atmospheric deposition of P (dry and wet deposition) was therefore <8% of the total riverine P input. This percentage was comparable to ~10% of the riverine P reported for other locations in the world (Duce, 1986; Moutin et al., 1998). Efforts in improving water quality in Lake Taihu have been focused on reducing riverine P. When the riverine input is reduced, the relative importance of atmospheric P input will increase. Our estimates of atmospheric input of P derived from measurements of atmospheric P, although coarse in nature, indicate that this source of P will need to be considered in the future.

4. Conclusion

The P concentration in total suspended particles ranged from 0.054 to 0.300 μg m^{-3} in the northern part of Lake Taihu. Soluble inorganic phosphorus, which offers the direct nutrition sources to the harmful algae, accounted for 4.9–83.4% of the total aerosol phosphorus measured with the HNO$_3^-$ HF digestion method. Correlations between soluble inorganic phosphorus and EC, WSOC, NO$_3^-$ and Ca^{2+} and Principal Component Analysis suggest that the aerosol soluble inorganic P above Lake Taihu may mainly come from two nearby coal-fire power plants. The average concentration of the total P in rain water samples was 0.082 ± 0.058 mg P/L.

Atmospheric input of P to the northern part of Lake Taihu was estimated using the P measurements in aerosol and rainwater samples. The annual dry deposition flux of P was estimated to be 0.04 kg ha^{-1} year^{-1} corresponding to a lower limit deposition

Table 4
Estimation of dry deposition flux of aerosol phosphorus into Lake Taihu.

	Dry deposition flux (μg (m^2 d))	
	Lower estimate	Upper estimate
Winter	13	135
Spring	11	113
Summer	5	52
Autumn	18	178
Annual average	12	120

velocity of 0.1 cm s^{-1} or 0.44 kg ha^{-1} year^{-1}corresponding to an upper limit deposition velocity of 1.0 cm s^{-1}. The wet deposition input flux of P was estimated to be 0.33 kg ha^{-1} year^{-1}. In comparison with the riverine phosphorus input into the northern part of Lake Taihu, the atmospheric input of P made a minor contribution (<8%) to the total P input.

Acknowledgements

Appendix Supplementary. material

Supplementary data associated with this article can be found in the online version. Comparison of F^{-} in rainwater samples collected over Lake Taihu with those at other sites studied. Comparison of three methods to determine the total phosphorus. Back trajectories of air masses that arrived at the sampling site calculated by HYSPLIT in the first day of each sampling month.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2011.01.043.

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